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Mid-infrared ultrashort pulses of 9.2-μm center wavelength are characterized in both amplitude and phase. This is achieved by use of a variant of spectral phase interferometry for direct electric field reconstruction in which spectral interferometry has been replaced with time-domain interferometry, a technique that is well suited for infrared pulses. The setup permits simultaneous recording of the second-order interferometric autocorrelation, thus providing an independent check on the retrieved spectral phase.

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The complete characterization—in both amplitude and phase—of ultrashort pulses has given rise to a great variety of experimental techniques,[1–3] with the advent of frequency resolved optical gating (FROG),[2] spectral phase interferometry for direct electric field reconstruction (SPIDER),[4] and a homodyne optical technique for SPIDER (HOT SPIDER)[5] that improves the sensitivity of SPIDER through the use of homodyne detection. However, few such complete measurements of ultrashort pulses have been demonstrated in the mid-infrared (MIR) domain, one that is important for many applications such as vibrational femtosecond spectroscopy and coherent control of molecular vibrations. Full characterization at these wavelengths is hampered by absorption in conventional linear and nonlinear optical materials and by the high cost, low sensitivity, and undesirable noise properties of MIR detector arrays. When an appropriate reference pulse is available, full MIR characterization can be made with techniques such as cross-correlation FROG,[6] electro-optic sampling,[7] and SPIDER in an upconversion scheme.[4] However, self-referenced measurements have been reported so far only for wavelengths of 5 μm and shorter, with techniques such as FROG[8,9] and the use of frequency-resolved pump–probe experiments.[10]

In this Letter we report the complete characterization of MIR femtosecond pulses at a center wavelength of 9.2 μm. This characterization was achieved by use of time-domain HOT SPIDER,[11] a variant of frequency-domain HOT SPIDER[5] in which spectral interferometry is replaced with time-domain interferometry. Time-domain interferometry, also known as Fourier-transform spectroscopy,[12] is indeed well suited to the infrared domain, first because the longer infrared wavelength makes it easier to achieve interferometry in the infrared than in the visible, and second because no grating or detector array is required. Time-domain HOT SPIDER relies on the fact that the Fourier transform of a second-order interferometric autocorrelation trace yields a peak at twice the carrier frequency that corresponds to the spectrum of the frequency-doubled incident pulse.[13] As we show below, the interferometric second-order correlation between a replica of the incident pulse and the superposition of the incident pulse and of a stretched pulse thus yields the same information as frequency-domain HOT SPIDER. Time-domain HOT SPIDER has been reported recently for 800-nm pulses; the sequence of three pulses was generated by use of a pulse shaper.[11] This is demonstrated here, to our knowledge for the first time, by use of discrete optical components, which makes it suitable even at wavelengths at which pulse shapers are not available. In addition, this setup automatically provides redundant information that can be used to verify the experimental procedure and the retrieved spectral phase, as discussed below.

We generate our infrared pulses (tunable in the 5–13-μm range with pulse energies of 4–2 μJ) through difference-frequency mixing in a 0.5-mm GaSe crystal of the signal and idler beams produced by an optical parametric amplifier (repetition rate, 1 kHz).[14] These pulses are completely characterized in phase and amplitude by use of a surprisingly simple optical setup (Fig. 1) in conjunction with chopping and time-delay control. The experimental setup is a

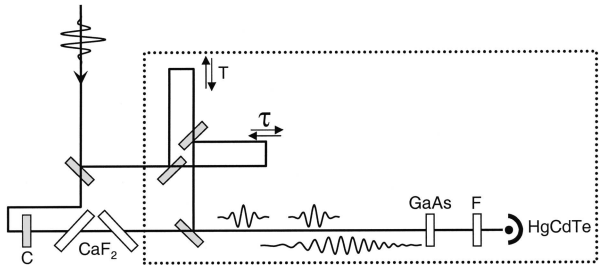

Fig. 1. Experimental setup for time-domain HOT SPIDER, based on a second-order collinear autocorrelator (area inside the dotted box). Second-harmonic generation is achieved in a GaAs crystal of 100-μm thickness and collected with a HgCdTe detector after transmission through a 2–6-μm bandpass filter (F). Scanning of time delay τ is achieved with a 2-Hz shaker and a copropagating He–Ne laser for accurate length calibration. To transform this standard autocorrelator into a time-domain HOT SPIDER apparatus we add two beam splitters, an optical chopper (C), and two tilted CaF_2 windows, thus generating the additional stretched pulse.

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standard second-order collinear autocorrelator, with one additional optical path in which a stretched pulse $E_S(t)$ is generated through linear dispersion in two tilted thick CaF_2 windows. Because of the huge dispersion of optical materials in the MIR domain, an effective thickness of only 34 mm results in a second-order phase of 162,000 fs^2, which is an appropriate value for our SPIDER measurement. The three-beam interferometer thus generates a sequence of three pulses: $E_S(t)$ and two replicas of the incident pulse, $E(t - \tau)$ and $E(t - T)$. After frequency doubling of the total electric field in a thin GaAs crystal, the signal recorded as a function of τ yields

$$S_T^{(2)}(\tau) = \int_{-\infty}^{+\infty} [E(t - \tau) + E(t - T) + E_S(t)]^4 \mathrm{d}t . \quad (1)$$

The stretched beam is mechanically chopped at half of the laser repetition rate; thus, when $E_S(t)$ is blocked, the standard second-order interferometric autocorrelation is measured. By subtracting the latter term from the signal measured when the stretched beam is not blocked, one obtains the differential correlation function, $\Delta S_T^{(2)}(\tau)$, associated only with the terms in Eq. (1) that depend on $E_S(t)$. This correlation function is shown in Fig. 2 for three values of delay T. Keeping only the terms in $\Delta S_T^{(2)}(\tau)$ that oscillate with respect to τ at twice the carrier frequency, we obtain

$$\Delta S_{T,2\omega}^{(2)}(\tau) = 6 \int_{-\infty}^{+\infty} \mathcal{E}^{*2}(t - \tau) \times [2\mathcal{E}(t - T)\mathcal{E}_S(t) + \mathcal{E}_S^2(t)] \mathrm{d}t , \quad (2)$$

where we have made use of the complex notation $E(t) = \mathcal{E}(t) + \mathcal{E}^*(t)$. Equation (2) corresponds to the correlation function between the frequency-doubled pulse, $\mathcal{E}^{(2)}(t) = \mathcal{E}(t)^2$, and the sum of (i) the sum-frequency mixing $\mathcal{E}_{\mathrm{SFM},T}(t)$ of $\mathcal{E}(t - T)$ with the stretched pulse and (ii) the frequency-doubled stretched pulse. The latter term is independent of T and thus contributes equally to the three measurements shown in Fig. 2. The data of Fig. 2(c), which correspond to a delay T such that only term (ii) contributes to the signal, can therefore be subtracted from those of Figs. 2(a) and 2(b) to provide term (i) of Eq. (2). Finally, a Fourier transform yields $\mathcal{E}^{(2)*}(\omega)\mathcal{E}_{\mathrm{SFM},T}(\omega)$, exactly as in frequency-domain HOT SPIDER except that this result was obtained through Fourier transforms of time-domain data. The last part of the calculation follows that of frequency-domain HOT SPIDER[5]: The instantaneous frequency of the stretched pulse is assumed to be constant during the overlap with $\mathcal{E}(t - T)$, so the spectral phase of $\mathcal{E}_{\mathrm{SFM},T}(\omega)$ is simply equal to $\varphi[\omega - \omega_S(T)] + \omega T$, where $\varphi(\omega)$ is the spectral phase that we want to measure and $\omega_S(t)$ is the instantaneous frequency of the stretched pulse, assumed to be governed by the independently measured dispersion of the CaF_2 window. Combining the measurements that correspond to Figs. 2(a) and 2(b) and subtracting $\omega(T_b - T_a)$, we obtain $[\omega_S(T_b) - \omega_S(T_a)](\mathrm{d}\varphi/\mathrm{d}\omega)$, from which the pulse's spectral phase can be retrieved through integration.

Figure 3 shows the spectral intensity and phase of our infrared pulses, indicating nearly transform-limited pulses of 105-fs FWHM duration. The spectral phase was retrieved by use of the time-domain HOT SPIDER procedure described above, whereas the spectral intensity was obtained directly through the Fourier transform of the first-order autocorrelation, thus yielding a better signal-to-noise ratio than the upconverted spectrum provided by SPIDER. We then repeated the measurement, using longer pulses obtained by inserting a 1.77-mm-thick CaF_2 window into the incident beam. The results, also shown in Fig. 3, correspond to negatively chirped pulses of 200-fs FWHM duration. The measured spectral phase is in excellent agreement with the sum of the known phase induced by the CaF_2 window and the measured phase of the nearly transform-limited pulses.

This experimental technique has the additional advantage that it automatically provides a method for independent verification of the spectral phase. The interferometric second-order autocorrelation functions that were subtracted out of the results earlier are shown in Fig. 4. For comparison, we show in Fig. 4 the interferometric second-order autocorrelation function calculated by use of the retrieved electric fields for both pulses [transform limited, Fig. 4(a); negatively chirped, Fig. 4(b)]. It is known that for a specific power spectrum and second-order interferometric autocorrelation function there is a unique spectral phase.[13] The excellent agreement observed in Fig. 4 is thus an independent check of the spectral phases shown in Fig. 3.

To summarize, we have shown that time-domain HOT SPIDER can be implemented through a simple

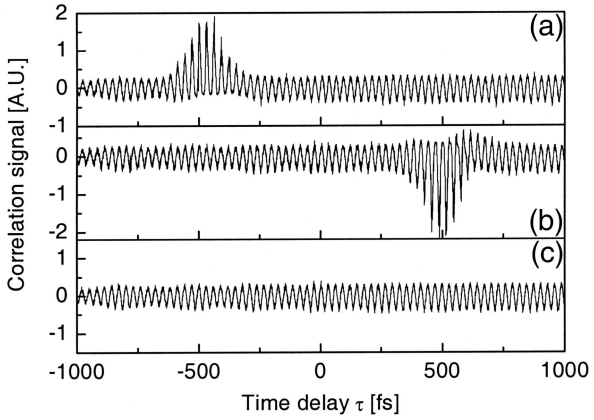

Fig. 2. Correlation function $\Delta S_T^{(2)}(\tau)$ measured for three values of delay T. $T_b - T_a = 990$ fs, corresponding to frequency shear $\delta\omega = 0.97$ THz. T_c is chosen such that the pulse replica $E(t - T_c)$ does not overlap the stretched pulse $E_S(t)$. Note that the fact that (a) points up and (b) points down is fortuitous: In (a) the short pulse $E(t - T)$ and the stretched pulse happen to interfere constructively during their overlap, whereas in (b) they interfere destructively. This effect depends on the exact choice of T and has no influence on the retrieved spectral phase.

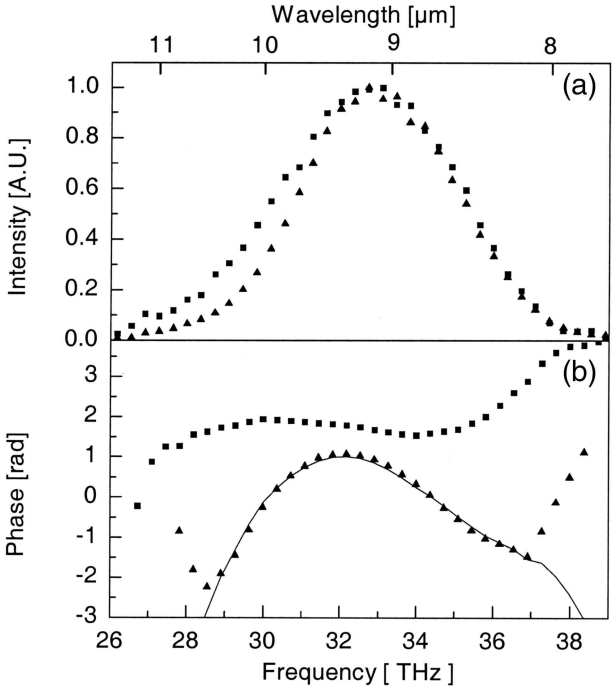

Fig. 3. Spectral (a) intensity and (b) phase of a nearly transform-limited infrared pulse (squares) and the same pulse after transmission through a 1.77-mm-thick CaF_2 window (triangles). Note the effect of absorption in CaF_2 on the low-energy side of the spectrum. The spectral intensity was obtained through a Fourier transform of the first-order autocorrelation that we recorded by blocking the stretched beam and removing the nonlinear crystal and filter in the setup of Fig. 1. The spectral phase was retrieved by use of the time-domain HOT SPIDER technique. The solid curve shows the spectral phase calculated by the addition of the known dispersion of CaF_2 to the phase measured for the undispersed pulse.

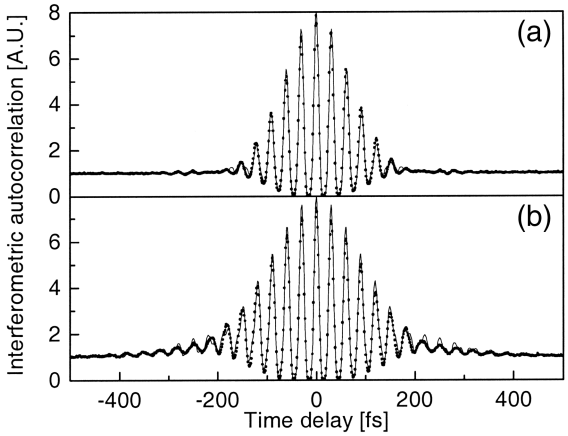

Fig. 4. Second-order interferometric autocorrelation (filled circles) measured (a) for the nearly transform-limited infrared pulses and (b) after dispersion through a 1.77-mm-thick CaF_2 window. The solid curves show a calculation based on the measured spectrum and the spectral phase retrieved by time-domain HOT SPIDER [Fig. 3(b)].

modification of a second-order collinear autocorrelator. Three differential correlation measurements are needed for retrieval of the spectral phase of the incident pulses. We applied this technique to mid-infrared pulses and achieved what we believe is the first complete self-characterization of femtosecond pulses near 10 μm. Time-domain HOT SPIDER shares with other SPIDER techniques the advantage of being a noniterative technique that relies on the measurement of data in only one dimension. It also possesses the general advantages of Fourier-transform spectroscopy,[12] such as the multiplex advantage (Fellgett advantage), the throughput advantage (Jacquinot advantage), easy choice of spectral resolution through the scanning range, and a broad spectral range determined mostly by that of the detector. The ability to increase spectral resolution easily is especially important for the measurement of complex pulse shapes. Although time-domain HOT SPIDER has the obvious drawback of not being single shot, it is an attractive technique for spectral domains such as the infrared for which detector arrays are not easily available. Furthermore, our experimental setup simultaneously provides the second-order interferometric autocorrelation as redundant information that can be used for checking the data's consistency, similarly to the well-known redundancy of a FROG trace.[2]

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